OB-17

The role of uncompensated electric charges in the polarization dynamics induced by femtosecond high-intensity infrared laser pulses

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Fast control over polarization is essential for many applications of ferroelectrics. Recently it was proposed to switch the polarization by an ultra-short laser pulse which frequency is in resonance with an infrared-active phonon mode Q_{IR} nonlinearly coupled to the ferroelectric soft mode Q_P [1]. The follow-up experiment [2] has indeed shown that a 150 fs midinfrared laser pulse with a fluence above 60 mJ cm^{-2} switches polarization in an irradiated part of $LiNbO_3$ crystal but then very fast, after about 0.2 ps, the polarization returns back to its initial value.

To model the soft mode dynamics in [1,2] a potential $V(Q_{IR}, Q_P)$ obtained ab-initio for different values of Q_{IR} and Q_P in unrelaxed lattice was used. We use instead the Landau thermodynamic potential $F(Q_{IR}, Q_P)$ which is crystal symmetry-invariant and known to describe well the soft mode dynamics [3]. Thus, we solve numerically the system of equations for two phonon coordinates:

$$\ddot{Q} + \gamma \dot{Q} + \partial F / \partial Q = 0 .$$

The solution of these equations for the parameters values as in the experiment [2] shows the polarization return, Figure 2, only if we take into account an electric field $\boldsymbol{E}$ due to uncompensated electric charges determined by the polarization difference ($\boldsymbol{P}_b - \boldsymbol{P}_d$), Figure 1a, which appear during the very fast switching.

We argue that uncompensated electric charges can also be responsible for the polarization which appears in quantum paraelectric $SrTiO_3$ [4] after minutes of repeating ultra-short midinfrared excitation pulses in the experimental conditions similar to [2]. These charges, Figure 1b, gradually grow when polarization appears just after the pulses due to induced strains and then they slowly recombine on a time scale of hours or much faster if illuminated by ultraviolet light.

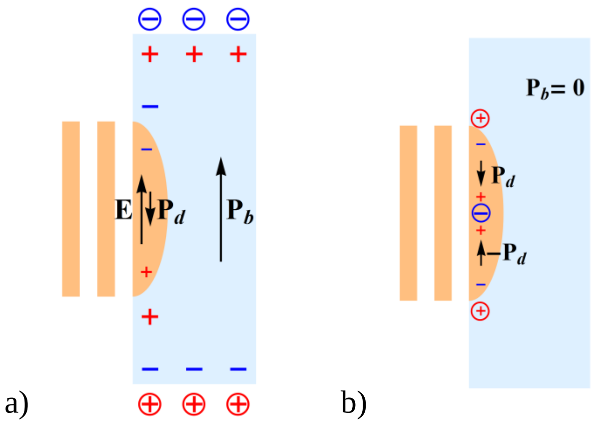

Figure 1. Scheme for free electric charges (in circles) and polarization-bound charges as in the experiments with (a) $LiNbO_3$ [2] and (b) $SrTiO_3$ [4].

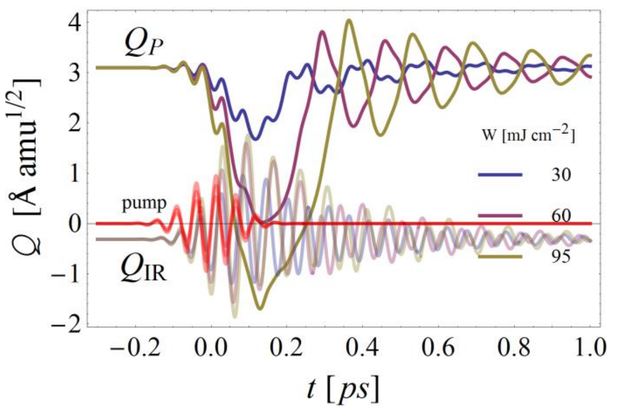

Figure 2. Numerical solution for the polarization dynamics in $LiNbO_3$ in conditions of the experiment [2].

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2. R. Mankowsky, A. von Hoegen, M. Först, A. Cavalleri, *Phys. Rev. Lett.* **118**, 197601 (2017).
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4. T.F. Nova, A.S. Disa, M. Fechner, A. Cavalleri, arXiv:1812.10560 (2018).